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SINGLE-CRYSTAL THERMIONIC CONVERTER STUDIES

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ABSTRACT

A thermionic converter is described in which the emitter is made of a single crystal of tungsten which has been cut to expose 16 square centimeters of 110 surface. Details of the cathode surface preparation, anode construction, and assembly techniques are presented along with some operating characteristics from the first successful run. The output power was not as large as expected. A second run following reprocessing demonstrated improved performance particularly at low cesium pressures.

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SINGLE-CRYSTAL THERMIONIC CONVERTER STUDIES*

H. F. Webster

I. INTRODUCTION

It has been known for some time that thermionic emission density from metals coated with a monolayer or less of cesium depends strongly on the nature of the underlying metal.[1-3] Thus, for example, when operated in the same pressure of cesium vapor, tungsten yields greater thermionic emission density than does niobium. This difference is reflected in thermionic converter performance and it is generally found that converters with tungsten cathodes yield higher output power densities than do ones with niobium cathodes. Recently, variations in thermionic emission density from different crystal faces of the same cesium-coated metal have been measured and found to be about as large as differences between metals.[3,4] Thus the exposed crystal face is a major variable, and this report will describe some studies made to control this variable and make use of it advantageously in thermionic converters.

The studies made with electron emission microscopes indicated that the crystal plane with the closest atomic packing is the one that will yield the maximum thermionic emission density in a given pressure of cesium vapor when the coverage is less than a monolayer. This close-packed plane would have the highest work function when in vacuum. The cathode of a thermionic converter is operated with coverage less than a monolayer, and thus the use of a controlled cathode surface made up preferentially of grains with close-packed atomic structures will allow operation of the device with minimum pressure of cesium vapor with the consequent minimum loss of output power in the resistance of the plasma.

The electron microscope studies have also indicated that in the region of more complete cesium coverage where minimum work function is obtained again, the work function is dependent upon the crystal structure of the base metal.[4] In this case, however, a simple rule for the choice of crystal face cannot be stated. It does appear, however, that the minimum work function face is frequently the atomically densest or second densest plane. Again, however, choice of a particular crystal plane for use as the anode surface of a thermionic converter will be advantageous.

II. CONTROL OF CRYSTAL SURFACE STRUCTURE

In order to accomplish the control of surface work function and cesium adsorption which would be desirable for thermionic converter operation, a variety of techniques may be used. These methods include mechanical

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deformation, heat treatment, and etching in various combinations. Basically, the problem has two parts: (1) control of the bulk grain structure, and (2) control of the actual crystal surface.

The bulk grain structure of refractory metal samples is usually determined by the mechanical forming and subsequent annealing processes. Thus, for example, rolling of sheet and drawing of wire frequently leave a particular orientation of grains statistically favored in the rolling and drawing direction. The statistics of grains exposed on the surface of the sheet and sides of the wire is thus established by the process. Subsequent annealing usually will lead to growth of larger grains particularly if the sample is free of impurities which tend to inhibit grain boundary motion. These larger grains will favor the more numerous seed grains if grain growth depends only on boundary curvature and not on crystal orientation. It has been shown, however, that grain growth in sheets of silicon-iron and tantalum[5] is strongly dependent upon the crystal orientation of the surface of exposed grains. Grains in the surface, which have high work-function surfaces exposed, grow at the expense of other grains. Thus after this surface energy growth process has operated, most of the surface will consist of high work-function grains even though these made up only a small part of the distribution before the anneal was carried out.

This particular grain growth process in tantalum not only leaves the proper grain structures, but the surface produced is such that minor deviations from the dense, high work-function planes are corrected out by terracing and the terrace planes are accurately 110 planes. No doubt similar grain growth processes occur in other metals, but these remain to be investigated.

The exposed crystal surface structure may also be controlled by chemical or electrolytic etching. These methods can be applied to materials with any basic grain structure from single-crystal forms to randomly oriented polycrystalline forms.

In the past few years large single crystals of refractory metals have become available which may be used to form converter electrodes. These may be cut to expose the correct crystalline face, but then the machined surface which usually consists of a mass of fine crystallites must be removed before a uniform high work-function plane will be left. This can be done in either of two ways. If the surface plane has been accurately ground to the desired crystal direction, then high voltage (i.e., $>$ 10 volts) electrolytic etching can be used. This has a tendency to remove material and to smooth a surface down to the average plane left by grinding. Probably a better method, however, which can be used if the dense, high work-function planes are to be left, is that of low voltage (i.e., 1 to 2 volts) electrolytic etching or chemical etching because a variety of these etches leave the dense planes preferentially. Thus, even if the grinding is a bit inaccurate, the etching procedure will correct out the error and leave the desired planes. Three methods of this sort which leave 110 planes on tungsten are low-voltage electrolytic etching in sodium hydroxide,[3] or chemical etching in nitric acid-hydrofluoric acid, or potassium ferricyanide. The surface left by the first procedure is shown in Fig. 1. It has also been found that aqua regia leaves 110 planes on vacuum-melted 18 per cent chromium iron alloy. Figure 2 shows the 110 planes left on the chrome iron by this

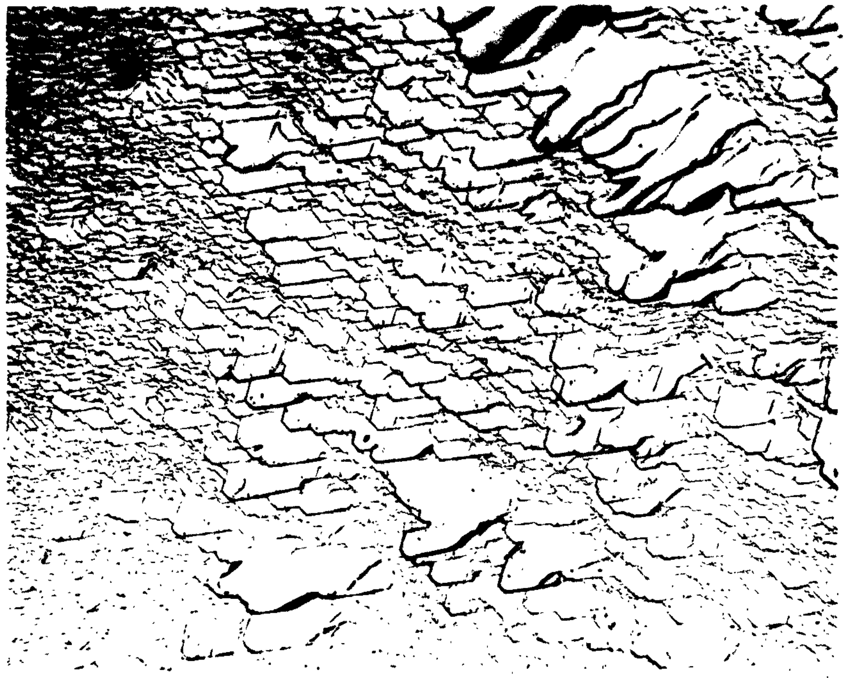

Fig. 1 Surface of tungsten single crystal following low-voltage electrolytic etching.

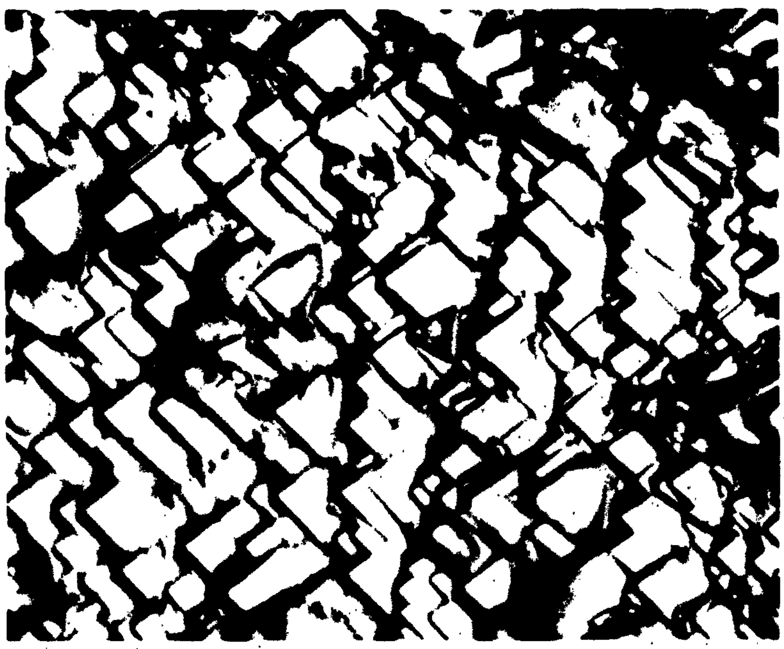

Fig. 2 Surface of etched chrome iron.

chemical etch. There are many other etching procedures which leave certain crystal faces preferentially and some of these are listed in Barrett, Structure of Metals.

III. THE "SINGLE-CRYSTAL" THERMIONIC CONVERTER

There are two ways in which control of the crystal structure of the electrodes may be beneficial in the operation of thermionic converters:

1. Choice of the optimum crystal face for either cathode or anode can be made in addition to choosing the best metal to use for each electrode. Thus the range of thermionic emission surfaces available to choose from is enlarged by the crystal surface variable.

2. Use of electrodes consisting mostly or entirely of the same crystal face will yield converter diodes of unusual uniformity since all parts will emit electrons or ions equally well. This is quite in contrast to present polycrystalline electrode diodes which have only a small fraction of their surface emitting electrons at maximum density. In addition, polycrystalline diodes will have most electrons from one type of grain and most ions some other type of grain and poor mixing will result. This problem of mixing of ions and electrons has been considered by Dobretsov[6] and by Houston and Webster.[7]

A problem related to the choice of crystal face to be used for the cathode which was suggested by M.D. Gibbons is illustrated in Fig. 3. This is a plot of the log of current density from two crystal faces immersed in cesium vapor vs reciprocal cathode temperature. Face 1 has a higher work function than face 2 when they are both hot enough to be free of cesium coverage. As has been found experimentally, face 1 then will yield the higher emission density at lower temperatures where partial cesium coverage occurs. It is clear then that a crossover must occur at a temperature designated as T_X. And, thus if the cathode temperature is less than T_X, face 1 should be used while face 2 should be used if above T_X. If surface ionization is the sole source of ions, the cathode temperature must be hot enough to supply sufficient ions to neutralize

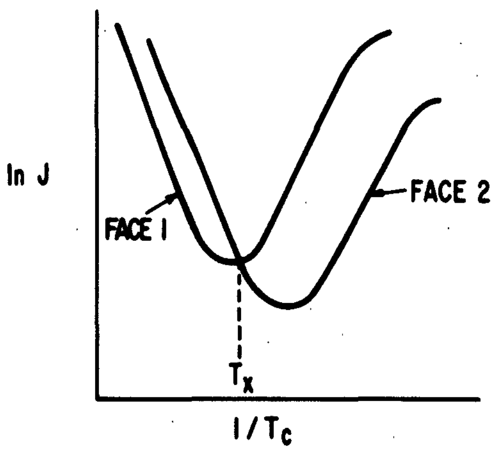

Fig. 3 Portions of emission S-curves for two crystal faces of same material.

the electron current. Ions generated by this process have been measured along with the electrons in the microscope studies for tantalum, tungsten, and rhenium, and for low cesium pressures; in each case it was found that sufficient ions were produced below T_X. It is not certain, however, that this will persist to high cesium temperatures. A complication to this problem is introduced by volume ionization which supplies an appreciable fraction of ions in efficient thermionic converters. This process will allow the cathode temperature to be lower than it would be if surface ionization were the sole source, and for these reasons the atomically dense, high work-function plane, face 1 appears to be the correct choice as cathode.

Some additional facts which must be considered when choosing the electrode surfaces are related to the thermal emissivities of the surfaces. For efficient converter operation, both cathode and anode should have the lowest possible thermal emissivities. The highly polished surfaces used in many conventional converters have quite low emissivity, and some sacrifice may be made in going to etched polycrystalline surfaces which may consist of many flat planes orientated at various angles. The single-crystal surfaces which have then been etched, such as is shown in Fig. 1, may, however, have quite low emissivities. Little exact information is presently available on the emissivities of single-crystal faces or geometrically rough surfaces left by etching.

IV. EXPERIMENTAL MODEL OF THE SINGLE-CRYSTAL THERMIONIC CONVERTER

A rectangular parallelepiped thermionic converter has been built that has a single-crystal tungsten cathode and an etched polycrystalline stainless steel anode. Details of its construction are shown in Fig. 4. Figures 5 and 6 are photographs of the cathode and anode sections taken before the tube was assembled. The cathode shown in Fig. 5 is of a square cross section with the round hole on its axis. This cathode configuration which was suggested by J. M. Houston has the 100 direction on its axis and the four major faces are all 110 planes. The bombardment heater coil is placed inside the tantalum tube and the tungsten crystal is then slipped over this tube. This has not been brazed in place, but good thermal contact is obtained because the expansion coefficient of tantalum is higher than that of tungsten. A thin section has been ground into the lower part of the tantalum tube which serves as the optimum current lead and the sole mechanical support for the cathode.

The anode section shown in Fig. 6 has a square hole on its axis, formed by electric discharge machining, which just fits around the cathode with the required cathode-anode spacing. The viewing holes visible at the four corners of the anode hole allow the cathode-anode gap to be viewed from outside the tube and thus to adjust the spacing after the tube is assembled. The bellows allowed motion of the cathode inside the anode hole and it could be fixed in various positions by use of the adjusting screws and support ring. This system was found to work very well and tubes have been assembled with average spacings of 0.005, 0.010, and 0.020 inch. In addition, the viewing holes permitted the cathode temperature to be monitored as well as visible discharge in the cathode-anode gap.

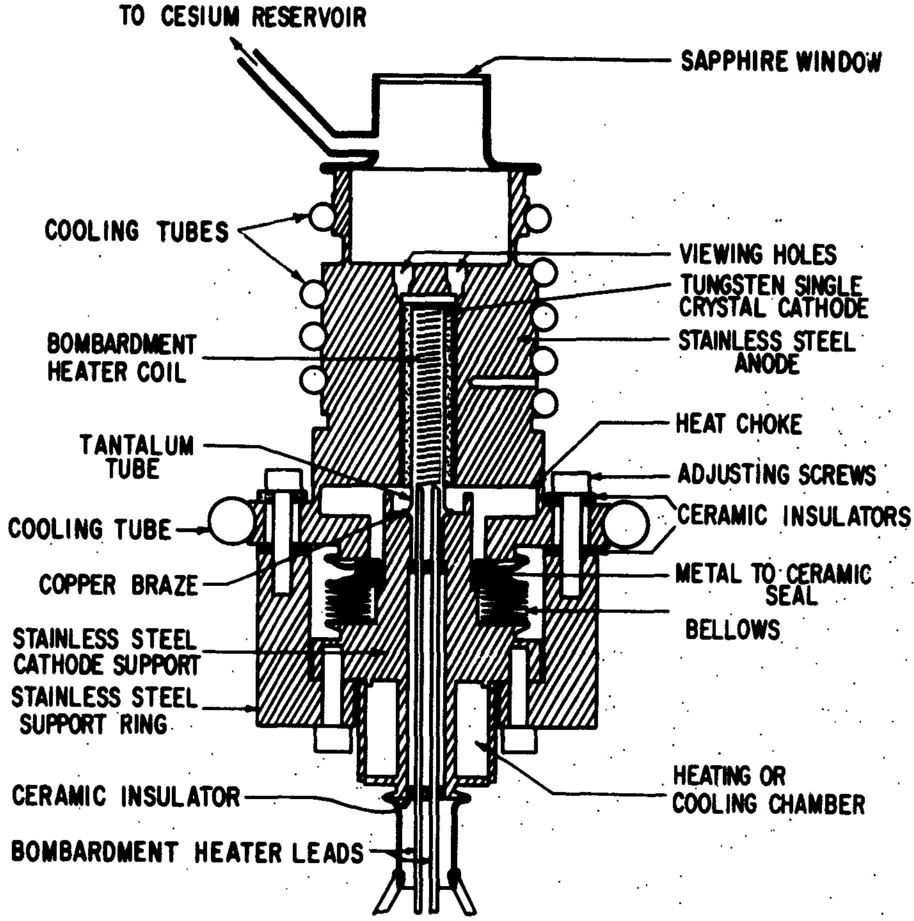

Fig. 4 Single-crystal cathode thermionic converter.

Fig. 5 Photograph of single-crystal cathode assembly.

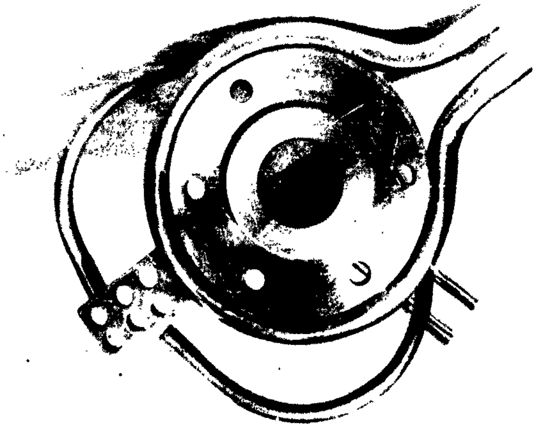

Fig. 6 Photograph of anode assembly.

The bombardment heater has been optimized to yield the maximum power density delivered to the cathode sleeve for a certain heater temperature by the procedure outlined by Houston, Howlett, and Webster. [8] The whole cathode-heater assembly is relatively efficient for several reasons:

1. The only conductive heat loss from the cathode is through the optimum lead,

2. The area of the hole through which the heater enters the cathode is a small fraction of the cathode area, and

3. The largest possible ratio of power delivered to the cathode by electron bombardment to that delivered by radiation has been attained.

A graph of cathode temperature vs input power required to attain that temperature is presented in Fig. 7. An advantage of such efficient cathode construction is that only a small correction will need to be applied to the measured converter efficiency to determine its true efficiency.

The converter is equipped with various cooling tubes through which air can be blown to control the temperatures of various parts of the tube. All parts of the tube were found to operate at suitable temperatures as a result of heat flows from the cathode with the exception of the window and tube to the cesium reservoir. These parts would fall below reservoir temperature unless auxiliary heating was applied.

The tube body has been made of stainless steel of one of three different compositions. These are commercial 347, commercial 430, or vacuum melted 430, which was prepared by the Research Laboratory's Metallurgy and Ceramics Research Department. The commercial 430 stainless steel contains certain deoxidants which unfortunately make this material difficult to arc weld. The vacuum melted 430, however, arc welds very well as also does the commercial 347. The 430 steel has two advantages for use in this tube over the 347 steel. The first advantage is that the 430 has a lower expansion coefficient than the 347, which more closely matches that of tantalum and thus makes the braze of

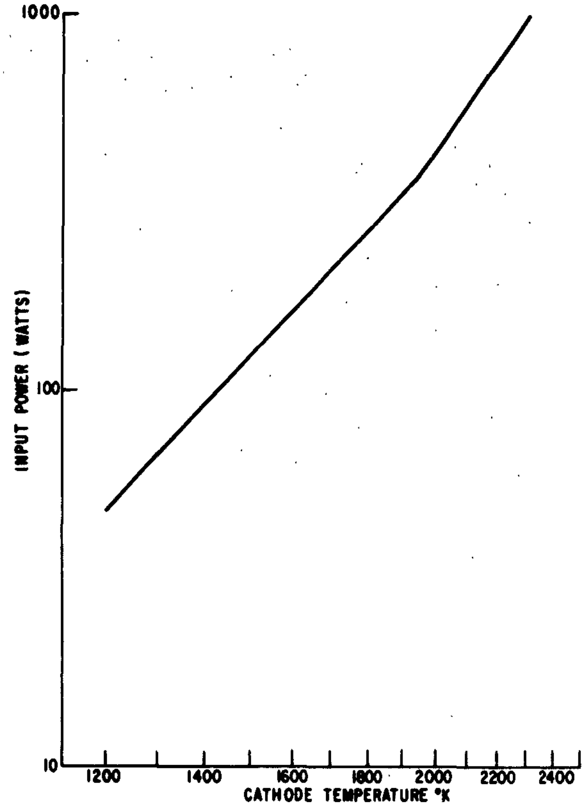

Fig. 7 Dependence of cathode temperature on input power with no electron cooling.

the tantalum tube to the cathode support less likely to leak when it is heat cycled. The second advantage of the 430 steel is that chemical etches have been found which leave high-density crystal planes on its surface. No such treatments have been found for the 347 stainless steel. With regard to the first advantage, it should be mentioned that three tantalum to 347 brazes have been made which have been heat cycled many times without failure. The geometry of these brazes, however, was as shown in Fig. 4 with a flared tantalum rim that avoids tearing the braze.

The sapphire window was brazed to a fernico spinning with silver solder, and this spinning was arc welded to the anode body. The metal-ceramic seal was made with high alumina ceramic rings and nickel disks by active alloy brazing, and this seal subassembly was connected to the bellows and anode section by arc welding. The various cooling tubes which are not exposed to cesium vapor were attached to the tube body by copper-gold brazes.

The separate vacuum chamber containing the bombardment heater coil was connected to an FN glass system with a Bayert-Alpert ion gauge and titanium getter bulb.

The cesium-containing volume of the tube was originally made to be free of any glass during operation. This was accomplished by distilling cesium into the converter and then pinching off a connecting nickel tube which then served as cesium reservoir. The tube has been operated twice with different cathode-anode spacings. The anode hole had a taper and the cathode-anode spacing for the first run was 0.009 to 0.014 inch when the tube was hot. The anode was made of commercial 430 stainless steel and an arc weld made to it could not be made leak-tight. The leak was sealed successfully with silicone resin and the tube pumped down to 10^{-9} torr. It was found, however, that cesium attacks the resin and a leak resulted after a brief period of operation.

The second run was made with the same anode, but it had been repaired by brazing on 347 stainless welding rings. The cathode-anode spacing was increased to 0.015 to 0.020 inch when the cathode became carburized in a brazing operation and the carbide layer was ground off and the cathode re-etched. The data presented in this report are from that run.

V. PERFORMANCE OF THE CONVERTER

The converter has been tested over a range of cathode temperatures from 1600° to 2100°K, of cesium reservoir temperatures from 528° to 640°K, and a variety of anode temperatures. A typical output current-voltage characteristic is shown in Fig. 8. It has been tested with two fillings of cesium, the first of which was contaminated with sodium which resulted from the decomposition of sodium hydroxide left over from the etching operation.

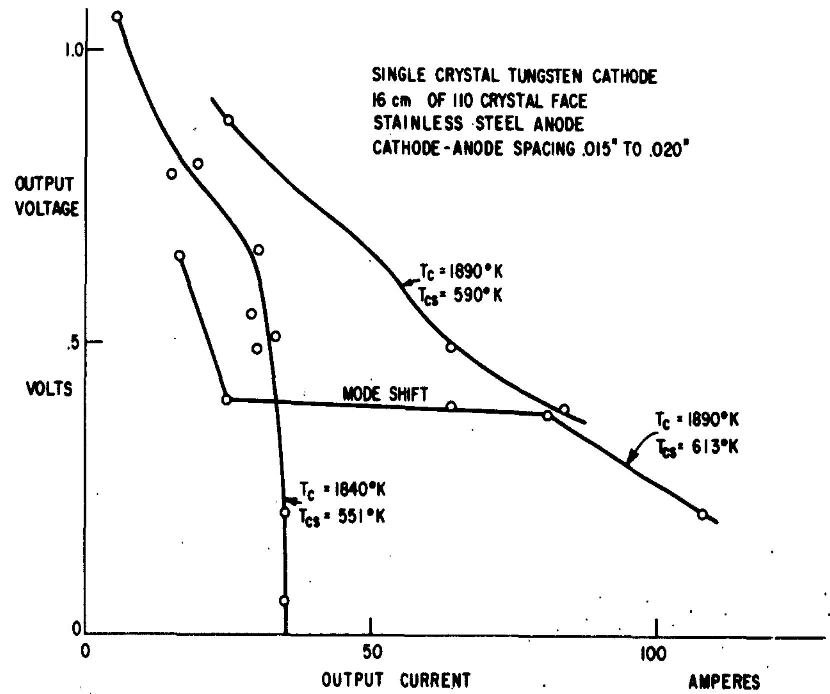

Fig. 8 Typical data showing output voltage as a function of output current.

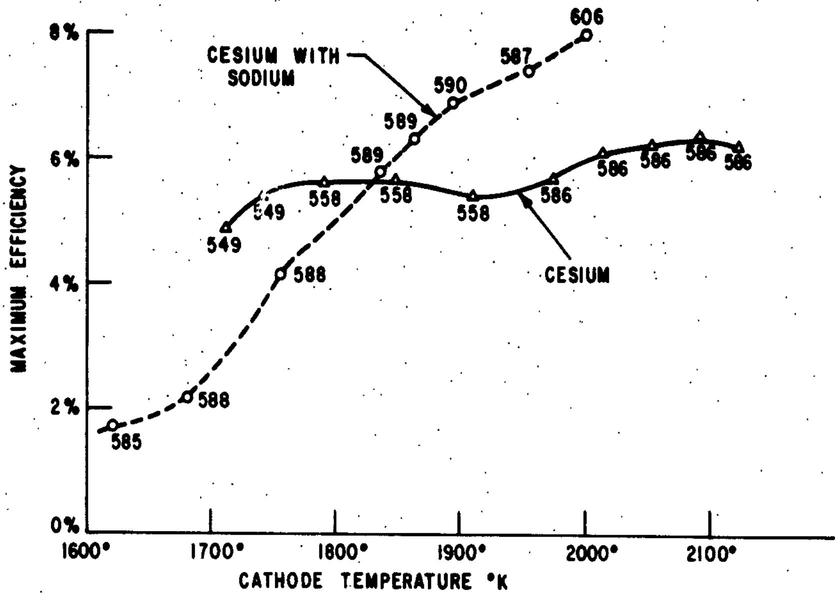

Fig. 9 Dependence of measured efficiency on cathode temperature and cesium pressure. Each point is labeled with the cesium bath temperature in °K.

Figure 9 shows the efficiency of the converter measured with the two cesium fillings as a function of cathode temperature. This value of the efficiency is not corrected for stray heat losses but is the total electrical power out over the total electrical power in. The first run made with the cesium-containing sodium had maximum efficiency values which increased from about 2 to 8 per cent over the cathode temperature range 1700° to 2000°K. After refilling the tube with clean cesium the device has almost constant efficiency and varied from about 5 to 6 per cent over the same temperature range. These values of efficiency were obtained with a lower cesium pressure than was used for the first run. Thus the performance was improved at low cesium pressures and low cathode temperatures, but was somewhat poorer at high cathode temperatures. The reason for this change is presently not understood.

It was recently found by Ausma Skerbele[9] that the addition of 1 per cent of sodium has a profound effect on the production of cesium ions in a heated tungsten surface. After the 1 per cent sodium was added it was necessary to raise the tungsten temperature by 375°K to obtain the same ion production that would have come from the tungsten if pure cesium was used. The sodium impurity could affect either the cathode or anode surface work function. It is not presently known how sodium interacts with the 110 face of tungsten.

There are two reasons why the efficiency of this converter may be low. The relatively wide cathode-anode spacing may introduce a plasma resistance effect. Wilson[10] has found that in high-pressure cesium converters the cathode-anode spacing is an important variable. He has found, for example, that in changing this spacing from 0.010 to 0.005 inch the output power density improves by a factor of two. Thus the spacing of 0.015 to 0.020 inch might be expected to degrade the performance below that of a 0.010-inch spaced converter.

In addition, the reflectivity of the etched commercial 430 stainless steel anode is unknown, but it is apparent from its matte grey appearance that its reflectivity is not as high as that of a polished anode surface.

It is planned to remake the anode of vacuum melted 430 stainless steel to yield a cathode-anode spacing in the range 0.007 to 0.010 inch. In addition to helping the plasma resistance problem, this anode can be etched to leave high-density crystal planes to improve the anode work function. In addition, this anode should have a much lower emissivity than the present model because this etching leaves brilliantly reflecting facets on that material.

ACKNOWLEDGMENTS

This tube is the work of a great number of people who have contributed to its design, construction, and testing. T. A. Howlett has ably assisted in all phases of the project. J. M. Houston, V. C. Wilson, J. Lawrence, and P. L. Read have shared in the design work. Fabrication and assembly has been skillfully carried out by personnel of the Laboratory shops. Crystal orientation has been determined by L. M. Osika, and surface microscopy has been done by R. R. Russell. V. L. Stout has encouraged the project through his continuing interest. I would like to thank all of them for their valued contributions.

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